# **REMARKS**

Claims 1-7 are pending in this application, of which claim 7 has been withdrawn from consideration and claims 1-6 have been amended. No new claims have been added.

A Substitute Specification correcting various grammatical, idiomatic and spelling errors is attached hereto, along with a marked-up version of the originally filed specification. No new matter has been added.

The Examiner has objected to claims 3 and 6 for various informalities which have been corrected in the aforementioned amendments. The title and the preamble of claims 1-6 have been amended to be directed to a <u>Gas Diffusion Layer Arrangement For a Fuel Cell</u>, because claim 2 recites a second gas diffusion layer stacked to what must be a first gas diffusion layer, both of them forming a gas diffusion layer <u>arrangement</u> for a fuel cell.

Figs. 1, 2 and 6 have been corrected. Figs. 1(a) and 1(b) have been labeled separately; Figs. 2(a) and 2(b) have also been labeled separately; and Fig. 6 has been labeled as "Prior Art".

Claim 4 stands rejected under 35 USC §112, second paragraph, as indefinite.

Accordingly, claims 1-6 have been amended to correct the noted instances of indefiniteness, and the 35 USC §112, second paragraph, rejection should be withdrawn.

Claim 1 stands rejected under 35 USC §102(b) as anticipated by or, in the alternative, under 35 USC §103(a) as unpatentable over U.S. Patent 3,793,085 to Hino et al. (hereinafter "Hino et al.").

Applicants respectfully traverse this rejection.

Hino et al. discloses a gas diffusion electrode for a fuel cell in which a layer 202 consisting of carbon powder subjected to a water repelling treatment surrounds an imbedded nickel net layer 203. Figs. 3 and 4 of Hino et al. show the layer 202 is present on both sides of nickel net layer 203.

This is in contrast to the present invention, in which the mixture of electrically conductive powder and water repellant (22) is contained entirely within the voids of mesh sheet (2), as shown in Fig. 1.

Accordingly, claim 1 has been amended to recite this distinction.

It should be noted that in the cell structure recited in claim 1 of the instant application, the air electrode comprises at least one gas diffusion layer and an air electrode side catalyst layer and the fuel electrode comprises at least one gas diffusion layer and a fuel electrode side catalyst layer, wherein an electrode reaction is carried out not at the gas diffusion layers, but at the catalyst layers. In other words, a fuel electrode reaction is carried out at a fuel electrode side catalyst layer and an air electrode reaction is carried out at an air electrode side catalyst layer (see page 2, lines 14 to 24 of the specification). The first and second gas diffusion layers function to

diffuse air and fuel to the fuel electrode side catalyst layer and the air electrode side catalyst layer, respectively.

The gas diffusion layer for the fuel cell recited in claim 1 of the instant application is excellent in gas permeability, water repellence or other, and has a remarkable effect of being able to supply the catalyst layer with reaction gas (air and fuel) by well diffusing, and at the same time, to discharge reaction produced water and removing water satisfactory. (See page 15, lines 4 to 8 of the specification).

Hino et al. discloses a gas diffusion electrode for a fuel call in which a layer (202) consisting of carbon powder and a binder thereof has a current collector (203) of a nickel net embedded therein (see claim 5 of Hino et al.). Hino et al. does not disclose the electrode consisting of a catalyst layer and at least one gas fusion layer, and in Hino et al. the layer (202) participates in an electrode reaction (see claim 5 of Hino et al.).

The advantageous results of the present invention as recited in claim 1 are shown at page 15, lines 4 to 8, of the specification. Those results are different from those of <u>Hino et al.</u>

Claims 2-4 stand rejected under 35 USC §103(a) as unpatentable over <u>Hino et al.</u> in view of Muranaka et al. (hereinafter "Muranaka et al.").

Applicants respectfully traverse this rejection.

Thus, the 35 USC §102(b) rejection should be withdrawn.

Muranaka et al. has been cited for teaching a three-layer electrode [56] on which the outer gas diffusion layer [52] is desired to have a lower wettability compared to the layer adjacent

the current collector [55], but neither of the cited references teaches either the limitations in claim 1, amended as proposed, from which these claims depend, or that the void rate of one layer is smaller than the other layer, as recited in claim 2 of the instant application.

It should be noted that in claims 2 to 4 of the instant application, a second gas diffusion layer is formed of the mixture of electrically conductive powder and water repellent filler.

The Examiner states: "<u>Hino et al.</u> teaches a second gas diffusion [305] stacked on the first gas diffusion layer [301] (Figure 5, line 5-13)" and "The first and second gas diffusion layers are both made of the Neoflon polyflon paper, thus the second gas diffusion layer is formed of the mixture of electrically conductive carbon powder and water repellant".

However, the Examiner's assertions are not in conformity with the following description found in column 5, lines 5-14 of **Hino et al.**:

An electrode of the present embodiment is shown in FIGS. 5 and 6 and a layer 305 consisting of powder of a fluorinated ethylene-propylene fluorocarbon resin, that is, "Neoflon", a polyflon paper 301 similar to that Embodiment IV, a layer 302 consisting of a mixture of activated carbon powder and fluorinated ethylene-propylene fluorocarbon resin powder, that is, "Neoflon", a current collector 303 in the form of a nickel net, and terminal 304.

If, according to the statement of the Examiner, the second gas diffusion layer is [305] and the first gas diffusion layer is [301], however, because the fluorinated ethylene-propylene fluorocarbon resin of [305] and the polyflon paper of [301] are not electrically conductive material, [301] and [305] can not be deemed to be the first and second gas diffusion layers which contain electrically conductive powder.

If Applicants could suppose that the Examiner intended to consider 302 as the first gas diffusion layer, the layers [301] and [305], which contain no electrically conductive powder, can not be deemed as the second gas diffusion layer, which contains electrically conductive powder.

In claim 2 of the instant application, the fuel cell has the arrangement of (air side) first gas diffusion layer/second gas diffusion layer/air side catalyst layer/electrolyte film/fuel side catalyst layer/second gas diffusion layer/first gas diffusion layer (fuel side), and a void rate of the second gas diffusion layer is smaller than that of the first gas diffusion layer, that is, the void rate of electrolyte side is smaller.

The Examiner states the Muranaka et al. teaches a three-layered electrode [56] on which the outer gas diffusion layer [52] is desired to have a lower wettability compared to the layer adjacent the current collector [55]. However, the layers [52]-[54] are catalyst layers (col. 8, line 20) and the catalyst layer has a gradient in water repellence across the thickness of the electrode that the water repellence is highest (highest void rate) in the area adjacent to the electrolyte membrane and lowest (lowest void rate) in the area adjacent to the conductor (dol. 10, lines 33-37 of Muranaka et al.). That is to say the highest void rate is on the electrolyte side. In contrast, in the present invention the void rate of electrolyte side is smaller and catalyst layers do not have such a gradient.

Therefore, <u>Muranaka et al.</u> does not disclose the features of claim 2 of the instant application.

The advantageous results obtained by claims 2 to 4 are shown at page 15, line 9 to page 16, line 4, of the specification. Those results are different from those of <u>Hino et al.</u> and

Thus, the 35 USC §103(a) rejection should be withdrawn.

Claims 5 and 6 under 35 USC §103(a) as unpatentable over <u>Hino et al.</u> in view of U.S. Patent 4,301,218 to <u>Benczur-ürmössy</u> (hereinafter "<u>Benczur-ürmössy</u>").

Applicants respectfully traverse this rejection.

Muranaka et al.

The Examiner has cited **Benczur-ürmössy** for teaching a second gas diffusion layer [1] having a thickness smaller than a first gas diffusion layer [3], in which the surface area of the first gas diffusion layer is smaller than that of the second gas diffusion layer.

Applicants respectfully disagree. Column 3, lines 1-17, cited by the Examiner, discloses neither the surface area nor the thickness of either the gas transport layer 1 or the working layer 4.

It should be noted that **Benczur-ürmössy** relates to a bi-porous electrode, including at least one conducting first layer having relatively large pores and at least one conducting second layer having relatively small pores to face a counter electrode in a cell, and containing Raneynickel (see claim 1). The gas transport layer 1 have large pores and fibers 2, and all the space 3 between the fibers serves for transporting gas. The working layer 4 includes the fibers 2 of the

substrate as well as the catalyst grains 5 (col. 3, lines 13-15). **Benczur-ürmössy**, however, does not disclose the first and second gas diffusion layers containing a electrically conductive powder and water repellent filler.

In the present invention, the gas diffusion layers do not comprise a catalyst.

The advantageous results obtained by claims 5 and 6 are shown at page 16, line 5 to page 19 of the specification. These results are different from those of **Benczur-ürmössy**.

Thus, the 35 USC §103(a) rejection should be withdrawn.

In view of the aforementioned amendments and accompanying remarks, claims 1-6, as amended, are in condition for allowance, which action, at an early date, is requested.

If, for any reason, it is felt that this application is not now in condition for allowance, the Examiner is requested to contact Applicants' undersigned attorney at the telephone number indicated below to arrange for an interview to expedite the disposition of this case.

U.S. Patent Application Serial No. 09/998,912 Response to Office Action dated May 19, 2004

In the event that this paper is not timely filed, Applicants respectfully petition for an appropriate extension of time. Please charge any fees for such an extension of time and any other fees which may be due with respect to this paper, to Deposit Account No. 01-2340.

Respectfully submitted,

ARMSTRONG, KRATZ, QUINTOS, HANSON & BROOKS, LLP

Welly Benle

William L. Brooks Attorney for Applicant Reg. No. 34,129

WLB/nrp Atty. Docket No. **011632** Suite 1000 1725 K Street, N.W. Washington, D.C. 20006 (202) 659-2930

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PATENT TRADEMARK OFFICE

Enclosures:

Substitute Specification w/Marked-Up Version

Abstract

Drawings (2 sheets)

Petition for Extension of Time

Q:\FLOATERS\WLB\01\011632\Amendment 05-19-04 OA



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### **SPECIFICATION**

#### TITLE OF THE INVENTION

GAS DIFFUSION LAYER <u>ARRANGEMENT</u> FOR <u>A</u> FUEL CELL <del>AND</del>

MANUFACTURING METHOD OF THE SAME

#### **BACKGROUND OF THE INVENTION**

1. Field of the Invention

The present invention concerns is directed to a gas diffusion layer 10 arrangement for a fuel cell and a manufacturing method of the same.

2. Detailed Description of the [[Prior]] Related Art

Fig. 6 is a dismantled cross-section an exploded cross-sectional view showing a basic configuration of an element cell of a solid polymer type fuel cell which is one embodiment of the conventional fuel cell. A cell is constructed by bonding an air electrode (cathode) side catalyst layer 2 containing a noble metal (mainly platinum) and a fuel electrode (anode) side catalyst layer 3 respectively to main faces at the both sides of a solid polymer electrolyte film 1. An air electrode side gas diffusion layer 4 and a fuel electrode side gas diffusion layer 5 are disposed respectively in opposition to the air electrode side catalyst layer 2 and the fuel electrode side catalyst layer 3. Thereby, an air electrode 6 and a fuel electrode 7 are configured respectively. These gas diffusion layers 4 and 5 [[has a]] function to pass an oxidant gas and a fuel gas, respectively, and, at the same time, to make the current flow to the outside. Then, an element cell 11 is configured by providing a gas passage 8 for reaction gas communication facing the cell, and pinching with a set of separators 10 provided with a cooling water passage 9 for cooling water communication on the opposed main faces and formed of an electrically conductive and gas impermeable material.

Fig. 7 is a cross-section view showing a basic composition of a solid polymer type fuel cell stack. A number of element cells 11 are stacked, sandwiched by a collector plate 12, an insulator plate 13 for of electric insulation and heat insulation, and a tightening plate 14 for maintaining the stacked state by applying a load, and tightened by means of bolts 15 and nuts 17 a tightening load being applied by a plate spring 16.

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The solid polymer electrolyte film 1 has a proton exchange group [[in]]  $\underline{at}$  the molecular <u>level</u>, and functions as  $\underline{a}$  proton electrically conductive electrolyte, as the specific resistance becomes equal to or less than 20  $\Omega$  cm2, if the water content is saturated. Thus, [[as]] <u>because</u> the solid polymer electrolyte film 1 functions as  $\underline{a}$  proton electrically conductive electrolyte by containing water, in the solid polymer type fuel cell, a method to operate by supplying each element cell 11 with reaction gas saturated with water vapor is adopted.

When the fuel electrode 7 is supplied with a fuel gas containing hydrogen, and the air electrode 6 is supplied with an oxidant gas containing oxygen, the fuel [[sell]] cell electrode reaction for decomposing hydrogen molecular into hydrogen ions and electrons takes place in the fuel electrode 7 and the following electric chemical reaction occurs for generating water from oxygen, hydrogen ions and electrons in the air electrode 6, respectively, where the load is supplied with power by electrons moving in an external circuit from the fuel electrode towards the air electrode, resulting in the production of water at the air electrode side.

Fuel electrode; 
$$H_2 \rightarrow 2H^+ + 2e^-$$
 (fuel electrode reaction)

Air electrode;  $2H^+ + (1/2) O_2 + 2e^- \rightarrow H_2O$  (air electrode reaction)

Whole;  $H_2 + (1/2) O_2 + \rightarrow H_2O$ 

Thus, in addition to water produced by the reaction at the air electrode 6 side, water moving from the fuel electrode 7 side to the air electrode 6 side

along with the movement of hydrogen ions also results [[in]].

Therefore, the gas diffusion layers 4 and 5 are required to assure the functions of 1) supplying the catalyst layer evenly with reaction gas to be supplied, 2) conducting the current to the outside, 3) controlling satisfactorily the supply/discharge of reaction produced water and moving water, or other functions.

Consequently, in the prior art, as gas diffusion layer 4 and 5, carbon paper, carbon cloth or other electrically conductive porous material, the electrically conductive porous material subjected to the water repellent treatment, or material coated with a mixture formed of carbon powder and water repellent filler on the electrically conductive porous material have been used.

However, the conventional gas diffusion layer [[was]] <u>is</u> expensive and, moreover, in case of carbon paper, it has been manufactured [[by]] <u>in</u> batch [[as]] <u>because</u> its mechanical strength is <del>not sufficient and fragile low,</del> making difficult to form continuously [[in]] <u>with</u> respect [[of]] <u>to</u> the electrode manufacturing, [[and]] <u>it</u> resulting in [[a]]poor productivity.

#### SUMMARY OF THE INVENTION

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The first object of the present invention is to solve the problems of the prior art and to supply a cheap inexpensive gas diffusion layer arrangement for a fuel cell excellent in water repellency, and which is also excellent in mechanical strength, allowing a continuous formation.

The second object of the present invention is to provide a method for manufacturing easily such a gas diffusion layer <u>arrangement</u> for fuel cell.

The <u>Inventors</u> have studied diligently in order to solve the problems of the prior art, and as a result, <u>have</u> found that the problems can be resolve <u>solved</u> by using a gas diffusion layer formed by using a mesh sheet

having [[an]] <u>a</u> heat resistance and an acid resistance such as, for example, stainless mesh, and filling voids of the sheet with a mixture of electrically conductive powder, such as carbon powder, and water repellent filler, such as fluorine resin, and <u>has</u> devised the present invention.

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In short, the gas diffusion layer for <u>a</u> fuel cell of claim 1 of the present invention is a gas diffusion layer used for at least one of gas diffusion layers of a fuel cell where a fuel electrode side catalyst layer and an air electrode side catalyst layer are disposed at both faces of an electrolyte film, and further a gas diffusion layer is disposed respectively on the outer surface of the fuel electrode side catalyst layer and air electrode side catalyst layer, characterized by that:

wherein the gas diffusion layer is formed of a mesh sheet having an heat resistance and an acid resistance, and a mixture of electrically conductive powder and water repellent filler for filling voids of the mesh sheet.

The gas diffusion layer for fuel cell of claim 2 is the gas diffusion layer for fuel cell of claim 1, wherein a A second gas diffusion layer [[is]] may be stacked on a face of the <u>first</u> gas diffusion layer in contact with the catalyst layer, the second gas diffusion layer being formed of the mixture of electrically conductive powder and water repellent filler, and presenting a void rate smaller than that of the <u>first</u> gas diffusion layer.

The gas diffusion layer for fuel cell of claim 3 is the gas diffusion layer for fuel cell of claim 2, wherein the The content of water repellent filler contained in the second gas diffusion layer is higher preferably greater than the content of water repellent filler contained in the <u>first</u> gas diffusion layer.

The gas diffusion layer for fuel cell of claim-4 is the diffusion layer for fuel cell of any of claims 1 to 3, wherein the fiber forming the mesh sheet is preferably coated beforehand with water repellent material.

The gas diffusion layer for fuel cell of claim 5 is the gas diffusion layer for

fuel cell of any of claims 2 to 4, wherein the thickness of the second gas diffusion layer is smaller preferably less than that of the first gas diffusion layer.

The gas diffusion layer for fuel cell of claim 6 is the gas diffusion layer for fuel cell of any of claims 2 to 5, wherein the electrically conductive powder used for the <u>first</u> gas diffusion layer and the second gas diffusion layer is <u>preferably</u> carbon powder, and the specific surface area of the carbon powder used for the <u>first</u> gas diffusion layer is <u>smaller preferably less</u> than the specific surface area of the carbon powder used for the second gas diffusion layer.

The claim 7 of the present invention is a manufacturing method of the gas diffusion layer for fuel cell of claim 1 or 2, comprising the A method of manufacturing the steps of; making a gas diffusion layer (precursor) using the mixture of electrically conductive powder, water repellent filler and hole making agent powder, or stacking further the second gas diffusion layer (precursor) and, thereafter, decomposing and scattering the hole making agent by heat treatment to form a gas diffusion layer having there fine holes.

#### BRIEF DESCRIPTION OF THE DRAWINGS

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- Fig.1 (a) is an illustrative drawing showing schematically the cross-section view of one embodiment of gas diffusion layer <u>arrangement</u> for <u>a</u> fuel cell of the present invention, while <u>Fig. 1(b)</u> is a plan illustrative drawing of a mesh and porous sheet having a heat resistance.
- Fig.2 (a) is an illustrative drawing showing schematically the eross-section cross-sectional view of another embodiment of <u>a</u> gas diffusion layer <u>arrangement</u> for <u>a</u> fuel cell of the present invention, while (b) is a plan illustrative drawing of a mesh sheet having a heat resistance and an acid resistance.
  - Fig. 3 an illustrative drawing showing the manufacturing method of one

embodiment of  $\underline{a}$  gas diffusion layer  $\underline{arrangement}$  for  $\underline{a}$  fuel cell of the present invention.

- Fig. 4 a graphic graph showing the relation between cell voltage current density.
- Fig. 5 a graphic graph showing the relation between cell voltage air availability.
  - Fig. 6 a dismantle cross-section an exploded cross-sectional view showing a basic configuration of an element cell of solid polymer type fuel cell which is one embodiment of the fuel cell.
  - Fig. 7 a cross-section cross-sectional view showing a basic composition of a solid polymer type fuel cell stack.

# **DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT**

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Now embodiments according to the present invention will be described in detail referring to the attached drawings.

- Fig. 1 (a) is an illustrative drawing showing schematically the cross-section of first embodiment of <u>a</u> gas diffusion layer <u>arrangement</u> for <u>a</u> fuel cell of the present invention, while <u>Fig.1(b)</u> is a plan illustrative drawing of a mesh and porous sheet having a heat resistance.
- In Fig. 1 (a) and <u>Fig. 1(b)</u>, it should be appreciated that the same components as components shown in Fig. 6 shall be referenced with the same symbols, and their duplicate <u>explanation shall</u> explanations will be omitted.

As shown in Fig. 1 (a), a gas diffusion layer 4 for <u>a</u> fuel cell of the present invention is formed by filling voids 21 of a mesh sheet 20 having a heat resistance and an acid resistance as a metal mesh with a mixture 22 of electrically conductive powder such as carbon powder and water repellent filler such as fluorine. A catalyst layer 2 is formed evenly on the upper portion of the

gas diffusion layer for fuel cell 4.

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The material of the mesh sheet 20 having a heat resistance and an acid resistance used for the present invention is not particularly limited, and for example, metals, ceramics, glass, engineering plastics or the others can be used. As metal, to be more specific, [[fro]] for example, stainless steel base metals (SUS316, SUS304, or others), [[titan]] titanium or [[titan]] titanium alloys or others can be cited. The hole diameter of the mesh sheet 20 is also not particularly limited, but it is preferable to used those [[of]] on the order of about 70 to 95% in the porosity. The form of the mesh sheet 20 is also not particularly limited, and any of metal gauze form, plain weave form, textile cloth form, mesh form, punching metal form can be used.

The mesh sheet 20 having a high mechanical strength can play the role of cell support member. Being formed by filling voids 21 of the mesh sheet 20 with the mixture 22 of electrically conductive powder such as carbon powder and water repellent filler such as fluorine, it is excellent in gas permeability, water repellency or others, can supply the catalyst layer with reaction gas by diffusing sufficiently, and at the same time, can discharged discharge reaction produced water or moving water satisfactorily. Moreover, an excellent mechanical strength thereof allows to form continuously easily, and to provide a cheap easy continuous formation, thereby providing an inexpensive gas diffusion layer.

Though the mesh sheet 20 can be used as it is, it is preferable to coat beforehand the fibers of the mesh sheet 20 with water repellent material such as fluorine resin. Such preliminary coating increases the water repellency in the proximity of the fabric of the mesh sheet 20, improves and [[keeps]] maintains the gas permeability, and at the same time, the water repellent material acts as an adhesive between the fabric and the electrically conductive powder such as

carbon powder, and prevents conductive powder such as carbon powder from falling from the gas diffusion layer for fuel cell 4.

As water repellent filler, to be more specific, example polytetrafluoroethylene, perfluorocarbon sulfonic acid, tetrafluoroethylene-perfluoroalkylvinyl ether copolymer, tetrafluoroethylene-hexafluoropropylene copolymer, polychlorotrifluoroethylene, polyfluorovynilidene, polyfluorovinyl, tetrafluoroethylene-ethylene and copolymer or others can be cited.

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To be more specific, as electrically conductive powder, for example, carbon powder, graphite powder, carbon fiber powder, metal powder, metal fiber powder, metal plating ceramics can be cited.

Fig. 2(a) is an illustrative drawing showing schematically the cross-section of another embodiment of gas diffusion layer for fuel cell of the present invention, while <u>Fig. 2(b)</u> is a plan illustrative drawing of a mesh sheet having a heat resistance and an acid resistance.

In Fig. 2 (a) and <u>Fig. 2(b)</u>, it should be appreciated that the same components as components shown in Fig. 6 shall be referenced with the same symbols, and their duplicate explanation shall explanations will be omitted.

As shown in Fig. 2(a), in the gas diffusion layer <u>arrangement</u> 4A for <u>a</u> fuel cell of the present invention, the gas diffusion layer 4 is formed by filling voids 21 of a mesh sheet 20 having [[an]] <u>a</u> heat resistance and an acid resistance as a metal mesh with a mixture 22 of electrically conductive powder such as carbon powder and water repellent filler such as fluorine. A second gas diffusion layer 23 is formed of a mixture of electrically conductive powder and water repellent filler in the same composition ratio as the mixture 22 or in a different composition ratio thereof on a face of the gas diffusion layer 4 to be come into contact with the catalyst layer 2 of the gas diffusion layer 4. The second gas

diffusion layer 23 presenting a void rate smaller less than that of the gas diffusion layer 4 is stacked. The catalyst layer 2 is formed uniformly on the upper portion of the second gas diffusion layer 23.

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By adopting [[the]] this composition, in the gas diffusion layer arrangement for a fuel cell 4A of the present invention, the gas diffusion layer 4 having a mesh sheet 20 of high mechanical strength can play the role of cell support member, while the second gas diffusion layer 23 allows to from permits formation of a more uniform catalyst layer 2. Besides, [[since]] because the second gas diffusion layer 23 presenting a void rate smaller than that of the first gas diffusion layer 4 is disposed by stack stacked upon the first gas diffusion layer 4, the second gas diffusion layer 23 can play a role to control satisfactory satisfactorily control the supply/discharge of reaction produced water and moving water.

Though the content of water repellent material contained in the second gas diffusion layer 23 is not particularly limited, it is preferably higher greater than the content of water repellent material contained in the <u>first</u> gas diffusion layer 4. Evaporation and scattering of reaction produced water and moving water into the reaction gas can be controlled by increasing the content of water repellent material contained in the <u>first</u> gas diffusion layer 4.

Though the mesh sheet 20 can be used as it is, as mentioned above, it is preferable to coat beforehand the fabric of the mesh sheet 20 with a water repellent material such as fluorine resin, because such preliminary coating increases the water repellency in the proximity of the fabric of the mesh sheet 20, improves and keeps the gas permeability, and at the same time, the water repellent material acts as adhesive between the fabric and the electrically conductive powder such as carbon powder, and prevents conductive powder such as carbon powder from falling from the gas diffusion layer for fuel cell 4.

The thickness of the second gas diffusion layer 23 is not limited particularly. It is preferable that the thickness of the second gas diffusion layer 23 be inferior to <u>less than</u> the thickness of the gas diffusion layer 4. By doing this, the effect of the second gas diffusion layer 23 presenting a lower gas diffusion capacity can be limited, and the gas diffusion of the [[whole]] <u>entire</u> cell can be maintained.

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The electrically conductive powder used for the <u>first</u> gas diffusion layer 4 and the second gas diffusion layer 4A is not specified particularly. However, carbon powder, being [[cheap]] <u>inexpensive</u> and easily available, can be used preferably as electrically conductive powder. Moreover, it is preferable to eempose—so that the specific surface area of the carbon powder of the gas diffusion layer 4 be <u>smaller less</u> than the specific surface area of the carbon powder of the second gas diffusion layer [[4A]] <u>23</u>.

Whereby, the absorbency of the <u>first</u> gas diffusion layer 4 becomes higher <u>greater</u> than that of the second gas diffusion layer <u>23</u>, allowing to move excessive moisture in the second gas diffusion layer [[4A]] <u>23</u> and evaporate in the gas, without stagnation or detention.

Fig. 3 is an illustrative drawing showing the manufacturing method of one embodiment of gas diffusion layer for fuel cell of the present invention shown in Fig. 2.

As shown in Fig. 3, first, a mesh sheet having a heat resistance and a acid resistance as metal mesh is prepared, and then, treated with emulsion of water repellent material such as fluorine resin, and a water repellent treated mesh sheet is made by heat treatment.

Thereafter, a <u>first</u> gas diffusion layer (precursor) is formed by filling voids of the water repellent treated mesh sheet with a mixture of electrically conductive powder, water repellent filler and hole making agent powder. Then,

the second gas diffusion layer (precursor) is formed by applying and stacking the mixture of electrically conductive powder and water repellent filler on the <u>first</u> gas diffusion layer (precursor).

Though, in the aforementioned embodiment, the second gas diffusion layer (precursor) is heat treated after [[stack]] stacking, the second gas diffusion layer may also be formed after the heat treatment of the first gas diffusion layer (precursor) before stacking the second gas diffusion layer (precursor) for obtaining a gas diffusion layer by decomposing, scattering and removing the hole making agent.

By the manufacturing method, it becomes possible to manufacture easily the gas diffusion layer <u>arrangement</u> for <u>a</u> fuel cell of the present invention where the void rate of the gas diffusion layer <u>arrangement</u> is changed <del>as will</del> only by adding steps of decomposing, scattering and removing the hole making agent.

# 15 EMBODIMENT OR EXAMPLE

Now the present invention shall be described more in detail referring to examples and comparison examples; however, the present invention is not limited by these examples [[at all]].

### (Example 1)

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- 20 (1) A metal mesh made of SUS316 (line diameter 0.2 mm) is soaked in a FEP dispersion adjusted to the specific weight of 1.09 and thereafter, dried, and heat treated (360 °C, 30 min) to prepare a metal mesh having a FEP layer partially formed on the surface.
- (2) Vulcan XC-72 (specific surface area : 250 to 300 m3/g) 7g, PTFE
  25 (polytetrafluoro ethylene ) powder (PTFE 6CJ) 3g and hole making agent powder (ammonium acid carbonate) 14g are mixed using kerosene as dispersion medium, and excessive kerosene is removed. The obtained mixture

is formed into a sheet by rollers.

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- (3) The sheet mold obtained by the aforementioned (2) and the metal mesh obtained by the aforementioned (1) are stacked, calendered and thereby finished to a thickness substantially equal to the metal mesh thickness, and a sheet shape mold [first gas diffusion layer (precursor)] where voids of the metal mesh is filled with the aforementioned mixture is obtained.
- (4) Hole making agent powder (ammonium acid carbonate) is decomposed, scattered and removed through the heat treatment of the sheet shape mold obtained in the aforementioned (3) at 60 °C for 30 min, to prepare a <u>first</u> gas diffusion layer (precursor).
- (5) Vulcan XC-72 6g, 60 weight 5 of PTFE dispersion 6. 67g are mixed using terpionel as dispersion medium, to prepare a past having an appropriate viscosity.
- (6) The paste obtained in the aforementioned (5) is applied to the sheet [first gas diffusion layer (precursor)] obtained in the aforementioned (4) to a thickness of the order of 0.05 mm and, thereafter, dried at 60 °C for 30 min, and a second gas diffusion layer (precursor) is stacked on the first gas diffusion layer (precursor).
- (7) After drying, the gas diffusion layer <u>arrangement</u> for <u>a</u> fuel cell of the 20 present invention is made by sintering PTFE (water repellent filler) through heat treatment of 360 °C for 30 min.

### (Example 2)

- (1) A metal mesh made of SUS316 (line diameter 0.2 mm) is soaked in a FEP dispersion adjusted to the specific weight of 1.09 and thereafter, dried, and heat treated (360 °C, 30 min) to prepare a metal mesh having a FEP layer partially formed on the surface.
  - (2) Ketjenblack (specific surface area: 800 m3/g) 7g, PTFE powder

- (PTFE 6CJ) 3g and hole making agent powder (ammonium acid carbonate) 14g are mixed using kerosene as dispersion medium, and excessive kerosene is removed. The obtained mixture is formed into a sheet by rollers.
- (3) The sheet mold obtained by the aforementioned (2) and the metal mesh obtained by the aforementioned (1) are stacked, calendered and thereby finished to a thickness substantially equal to the metal mesh thickness, and a sheet shape mold [first gas diffusion layer (precursor)] where voids of the metal mesh [[is]] are filled with the aforementioned mixture is obtained.
- (4) Hole making agent powder (ammonium acid carbonate) in the sheet shape mold [first gas diffusion layer (precursor)] is decomposed, scattered and removed through the heat treatment of the sheet shape mold [first gas diffusion layer (precursor)]obtained in the aforementioned (3) at 360 °C for 30 min, after having dried at 60 °C for 30 min, and at the same time, PTFE (water repellent filler) is sintered to prepare a first gas diffusion layer.
- (5) Vulcan XC-72 6g, 60 weight % of PTFE dispersion 6. 67g are mixed using terpionel as dispersion medium, to prepare a past having an appropriate viscosity.
- (6) The paste obtained in the aforementioned (5) is applied to the sheet (<u>first</u> gas diffusion layer) obtained in the aforementioned (4) to a thickness of the order of 0.02 mm and, thereafter, dried at 60 °C for 30 min, and a second gas diffusion layer (precursor) is stacked.
- (7) After drying, the gas diffusion layer for fuel cell of the present invention is made by sintering PTFE (water repellent filler) in the second gas diffusion layer (precursor) through a heat treatment of 360 °C for 30 min.

# 25 (Comparison Example 1)

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(1) A carbon paper TGP-060 (thickness: 0.2 mm) made by TORAY Co., LTD. is soaked in a PTFE dispersion adjusted to a specific weight of 1.10 and

thereafter, dried, and heat treated (360 °C, 30 min).

- (2) Vulcan XC-72 6g, 60 weight % of PTFE dispersion 6. 67g are mixed using terpionel as dispersion medium, to prepare a past having an appropriate viscosity.
- (3) The paste obtained in the aforementioned (2) is applied to the sheet obtained in the aforementioned (1) to a thickness of the order of 0.02 mm and, thereafter, dried at 60 °C for 30 min, to make a gas diffusion layer.
- (4) After drying, the gas diffusion layer for comparison is formed by sintering PTFE (water repellent filler) through a heat treatment of 360 °C for 30 min.

# [Element Cell for Test]

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An element cell for <u>a</u> test of <del>25 cm2 in</del> <u>an</u> electrode area <u>of 25 cm²</u> is prepared for evaluation of respective cells, by making the gas diffusion layer <u>arrangement</u> prepared in the Examples 1 and 2 and Comparison Example 1 the air electrode side gas diffusion layer, and using the gas diffusion layer <u>arrangement</u> prepared in the Comparison Example 1 as the fuel electrode side gas diffusion layer <u>arrangement</u> for all.

### [Test Results]

- Fig. 4 shows the current voltage characteristics of the element cell using the gas diffusion layer <u>arrangement</u> of the Examples 1 and 2 and Comparison Example 1. In every element cell prepared by any of Examples, performances substantially equal to the comparison example 1 can be obtained.
- Fig. 5 shows the air availability dependency of cell voltage in each element cell of the Examples 1 and 2 and Comparison Example 1. In case of Example 2, the voltage tends to lower suddenly at the high air availability side, while the voltage drop amount at the lower air availability side tends to become smaller than that of the conventional example and the Example 1, whereby,

performances substantially equal to the comparison example 1 can be obtained.

The gas diffusion layer <u>arrangement</u> for <u>a</u> fuel cell of <u>claim</u> of the present invention being formed of a mesh sheet having [[an]] <u>a</u> heat resistance and an acid resistance, and a mixture of electrically conductive powder and water repellent filler for filling voids of the mesh sheet, it becomes possible to prepare the gas diffusion layer occupying a large part of the material cost of the element cell of fuel cell at a low cost, [[as]] <u>and</u> a high strength and [[cheap]] <u>inexpensive</u> material can be used as mesh sheet and, moreover, a production excellent in mass productivity is enabled as it becomes possible to take up the gas diffusion layer (sheet) by using a flexible material as <u>a</u> mesh sheet, and also the mesh sheet plays the role of cell support as it present a high mechanical strength, or other remarkable effects are deployed.

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Besides, the gas diffusion layer for fuel cell of claim 1 arrangement of the present invention is excellent in gas permeability[[,]] and water repellency or others, and deploys exhibits a remarkable effect of being able to supply the catalyst layer with reaction gas by well diffusing, and at the same time, to discharge reaction produced water and moving water satisfactory.

As the gas diffusion layer for fuel cell of claim 2 is the gas diffusion layer for fuel cell of claim 1, wherein a A second gas diffusion layer is preferably stacked on a face of the first gas diffusion layer in contact with the catalyst layer, the second gas diffusion layer being preferably formed of the mixture of electrically conductive powder and water repellent filler, and presenting a void rate smaller than that of the first gas diffusion layer, it can deploy the same effects as the gas diffusion layer for fuel cell of claim 1 and, furthermore Furthermore, the second gas diffusion layer allows to form formation of a more uniform catalyst layer, and moreover, [[to]] allows satisfactory control satisfactorily of the supply/discharge of reaction produced water and moving

water.

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The gas diffusion layer for fuel cell of claim 3 is the gas diffusion layer for fuel cell of claim 2, wherein the The content of water repellent filler contained in the second gas diffusion layer is preferably formed higher than the content of water repellent filler contained in the <u>first</u> gas diffusion layer and has a remarkable effect of allowing to control controlling the evaporation scattering of reaction water and moving water to the reaction gas.

The gas diffusion layer for fuel cell of claim 4 is the diffusion layer for fuel cell of any of claims 1 to 3, wherein the The fiber forming the mesh sheet is preferably coated beforehand with water repellent material and, thereby, has remarkable effects of enhancing enhances the water repellency in the vicinity of the fiber of mesh sheet, improving and keeping the gas permeability, and making the repellent material to act as adherent between the fiber and the electrically conductive powder as carbon powder, thereby preventing the electrically conductive powder as carbon powder from falling from the gas diffusion layer arrangement for a fuel cell.

The gas diffusion layer for fuel cell of claim 5 is the gas diffusion layer for fuel cell of any of claims 2 to 4, wherein the The thickness of the second gas diffusion layer is smaller less than that of the first gas diffusion layer, whereby, the effect of the second gas diffusion layer poor in gas diffusion is reduced, and has a remarkable effect of maintaining the gas diffusion of the whole cell.

The gas diffusion layer for fuel cell of claim 6 is the gas diffusion layer for fuel cell of any of claims 2 to 5, wherein the electrically conductive powder used for the <u>first</u> gas diffusion layer and the second gas diffusion layer is carbon powder, which is easily available and cheap, and the specific surface area of the carbon powder used for the <u>first</u> gas diffusion layer is made <u>smaller less</u> than the specific surface area of the carbon powder used for the second gas

diffusion layer, thereby, the absorbency of the <u>first</u> gas diffusion layer becomes higher <u>greater</u> than that of the second gas diffusion layer, <u>allowing to have a remarkable effect of allowing to move excessive excess</u> moisture in the second gas diffusion layer <u>to move</u> and evaporate in the gas, without stagnation or detention.

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The manufacturing method of the gas diffusion layer for <u>a</u> fuel cell ef claim 7 of the present invention has a remarkable effect of allowing to manufacture easily permitting easy manufacturing of a gas diffusion layer for fuel {{cell}} wherein the void rate of the gas diffusion layer is changed as will, only by adding the step of decomposing, scattering and evaporating the hole making agent.

# **IN THE DRAWINGS:**

Figs. 1(a), 1(b), 2(a), 2(b) and 6 have been amended. Replacement sheets are attached for Figs. 1(a), 1(b), 2(a), 2(b) and 6.